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LABORATORY METHOD TO MEASURE SEALING CAPACITY OF CAPROCKS

METODYKA WYKONYWANIA POMIARÓW LABORATORYJNYCH DO OKREŚLANIA WŁASNOŚCI USZCZELNIAJĄCYCH SKAŁ NADLEGŁYCH

Adoption of geological sequestration as one of the main options for reducing carbon emissions to the atmosphere resulted in the intensification of research on various aspects relating to such a solution. The choice of structures for the potential storages must be preceded by a detailed characterization process. In addition to the appropriate location, capacity and injectivity, the most important issue is the proper seal, preventing the migration of injected CO_2 into overburden, and ultimately to the atmosphere. The process of granting licenses for the injection of CO_2 will require to confirm the sealing capacity of caprock by means of laboratory tests.

In response to current trends and requirements in the laboratory of the Faculty of Drilling, Oil and Gas of AGH UST in Krakow the apparatus for direct examination of the sealing efficiency of poorly permeable rocks, acting as seal rocks, was created according to current standards. This article discusses the mechanism of capillary seal and basic laboratory methods for determining the capillary threshold pressure for CO_2. The authors present the results of preliminary tests on shale samples with different permeabilities.

Keywords: threshold pressure, breakthrough pressure, sealing efficiency, capillary sealing, CO_2 sequestration

Przyjęcie geologicznej sekwestracji jako jednej z głównych opcji redukcji emisji dwutlenku węgla do atmosfery spowodowało intensyfikację badań nad różnymi aspektami związanymi z takim rozwiązaniem. Wybór struktur przeznaczonych na potencjalne składowiska musi być poprzedzony szczegółową ich charakteryzacją. Oprócz odpowiedniej lokalizacji, pojemności struktury czy jej iniekcyjności, najważniejszą kwestią jest odpowiednie uszczelnienie, uniemożliwiające migrację zatłoczonego CO_2 do warstw nadległych, a w efekcie do atmosfery. Proces przyznawania pozwoleń na zatłaczanie CO_2 będzie wymagał potwierdzenia szczelności struktury na drodze badań laboratoryjnych.

W odpowiedzi na aktualne tendencje i wymagania w laboratorium Wydziału Wiertnictwa, Nafty i Gazu Akademii Górniczo-Hutniczej w Krakowie utworzono stanowisko badawcze pozwalające na bezpośrednie badanie własności uszczelniających skał słabo przepuszczalnych, występujących w roli skał

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uszczelniających, zgodne z aktualnymi standardami w tym zakresie. W niniejszym artykule omówiono mechanizm uszczelnienia kapilarnego oraz podstawowe metody laboratoryjnego określania ciśnienia progowego dla CO_2. Autorzy przedstawili również wyniki wstępnych badań przeprowadzonych na próbkach skał łupkowych o różnych przepuszczalnościach.

Słowa kluczowe: ciśnienie progowe, ciśnienie przebicia, właściwości uszczelniające, uszczelnienie kapilarne, sekwestracja CO_2

1. Introduction

In view of the increasing concentration of CO_2 in the atmosphere actions are taken to reduce emissions of this greenhouse gas and its contribution to the atmosphere. One of the broader issue is the concept of geological sequestration of CO_2 in depleted hydrocarbon reservoirs, aquifers and deep coal seams. This technology is based on knowledge and experience from the oil and gas industry.

Geological structure dedicated to the storage, in addition to the required capacity and injectivity must have a proper sealing efficiency. Under the rules of implementing the EU Directive on geological sequestration of CO_2, as well as other international standards and guidelines, the quality of the seal is the primary criterion for selection of the structure. The ability of the sealing rocks to retain injected CO_2 should be confirmed as part of the characterization of potential storage sites in the way of laboratory tests on cores.

The sealing capacity of a low permeable rock is provided by the capillary forces across the interface of the wetting phase (brine) in the seal rock, and the nonwetting phase (oil or gas) in the reservoir. Initially, measurements of the capillary breakthrough pressure for low-permeable rocks (seals) have been performed in the context of the characterization of hydrocarbon deposits (also for investigating possibilities of underground gas storage overpressurization) or waste storages (e.g. nuclear). However, the available results cannot be directly used as the breakthrough pressure of CO_2 in the geological sequestration process. Due to significant differences in surface tension between CH_4/brine or N_2/brine and CO_2/brine systems breakthrough pressure is expected to be lower for carbon dioxide. Besides, the presence of CO_2 can lead to changes in the rock wettability. Therefore, the direct measurement of the sealing properties of caprocks is the most appropriate solution for determining upper limit of injection pressure for CO_2 storage (Li et al., 2005).

Sealing capacity features essentially two parameters: displacement pressure, which begins infiltration of the gas phase into the initially water (brine) saturated pore system of the sealing rock, and the relative permeability to gas phase after the gas breakthrough (Hildenbrand et al., 2002). Relative permeability is a function of the gas/water saturation of the pore system, which is not easy to determine if fine-grained rocks. The effectiveness of capillary seal is determined through laboratory tests.

2. Capillary sealing mechanism

The primary and dominant transport mechanism in the migration of hydrocarbons, both in the porous rock matrix and a network of fractures, is the Darcy flow. In the case of single phase

flow in the Darcy equation, the absolute permeability k_{abs} relates flow rate q of the fluid with viscosity μ with the pressure gradient ∇p causing the flow.

$$q = A\frac{k_{abs}}{\mu}\nabla p$$

In the situation when in a porous medium, there are two (or more) immiscible phases, there will be a two-phase (multiphase) flow, accompanied by capillary effects. The capillary forces at the interface between the wetting phase (brine), which saturates the sealing rock, and nonwetting phase (gas, oil, supercritical CO2) accumulated in the reservoir determine the ability of a rock to stop nonwetting phase flow. Infiltration of the nonwetting phase into idealized cylindrical pore throat of radius r is possible only after exceeding the capillary pressure, expressed by the equation:

$$P_c = P_{nw} - P_w = \frac{2\sigma}{r}\cos\theta$$

where:

σ — interfacial tension of wetting/non-wetting fluids (IFT) [Nm^{-1}],
θ — contact angle [°],
r — pore throat diameter [m].

The capillary sealing mechanism in the pore throat of sealing rock is illustrated schematically in Fig. 1.

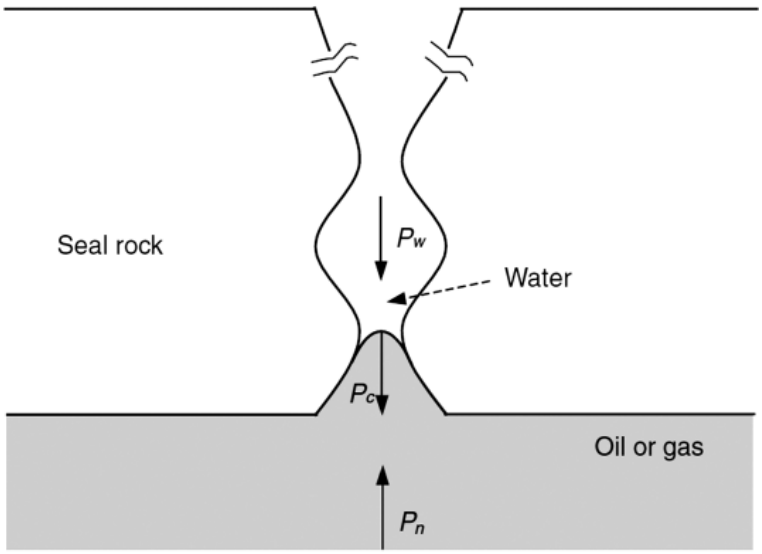


Fig. 1. Schematic of capillary sealing mechanism in a pore throat of seal rock (Li et al., 2005)

When the pressure difference between the nonwetting and wetting phase exceeds capillary pressure of given pore throat ($P_{nw} - P_w > P_c$), the nonwetting fluid will travel along the channel until it encounter channel of smaller diameter. In turn, when the differential pressure across the sealing rock exceeds capillary pressure of the interconnected pore system, creates a continuous "trickle" of nonwetting phase, resulting in slow Darcy flow. This pressure difference is defined as the breakthrough pressure.

3. Stages of capillary gas breakthrough

The nonwetting phase breakthrough process in the fine-grained porous medium consists of several stages, each corresponding to a characteristic value of the fluid pressure. At the beginning of the process nonwetting fluid fills only the largest pores on the surface of contact. This is possible when the pressure of injected fluid exceeds capillary entry pressure of the porous medium. Therefore, the excess of the first characteristic of the pressure starts nonwetting fluid migration into the porous rock sealing, but the flow in the entire cross section of the porous medium has not happened yet.

A further increase in nonwetting fluid pressure opens the next smaller pore throats, displacing the wetting phase and increasing nonwetting phase saturation in the pore system. During this phase, the gas pressure is higher than the capillary entry pressure, but still lower than the threshold pressure (breakthrough pressure) ($p_c^e < p_c < p_c^{th}$). Although there has been infiltration of nonwetting fluid into the pore system of seal rock, yet it retains its sealing properties. The next step is the creation of a continuous flow path along the entire cross-section of porous medium in the form of interconnected channels with the largest pore diameters. The pressure at which it formed a kind of gap in the capillary sealing system is called the capillary breakthrough pressure (threshold pressure) (Schowalter, 1979; Dullien, 1992; Hildenbrand et al., 2002, Li et al., 2005). At this stage flow (leakage) comprises a small part of the pore network. As the pressure increases the flow extends to the other pore throats, increasing the saturation of nonwetting phase and thus effective permeability for that phase.

It should be noted the difference between the capillary entry pressure and the breakthrough pressure. The first is a measure of the diameter of the largest pore on the surface of the rock sample under the given conditions of rock wettability and surface tension of fluids (Li et al., 2005). The second sets out the conditions under which it comes to leakage of stored fluid into the overburden.

The pressure reduction after nonwetting phase breakthrough will result in wetting phase re-imbibition starting with the smallest pore diameter. The gradual closing of flow paths decreases permeability for gas leading in effect to stop the flow (leakage)). An additional effect may be shut-off of the pores filled with gas from the rest of the pore system and the resulting residual nonwetting phase saturation.

4. Laboratory methods

1.1. Mercury porosimetry

The simplest and least time-consuming method of determining the threshold capillary pressure of rock samples is based on mercury porosimetry (Egermann et al., 2006). But simplicity is linked here with a number of limitations resulting from the assumptions and the capability of the mercury porosimetry itself. The main imperfection which may lead to significant differences between the results of the study and the reality is to assume a cylindrical geometry of the pore channels. Simultaneously, pore size range penetrated by mercury is limited by the maximum pressure achieved by the measuring apparatus, and therefore, the smallest pore size (radius < 2.1 nm, Schlömer & Krooss, 1997) may not be disclosed, what is especially important for fine-grained sealing rocks. Another issue that may affect the results obtained is the choice of method of sample preparation, and in principle its potential impact on the change of pore structure of the original system (Egermann et al., 2006).

Another major drawback of the method based on mercury porosimetry is lack of confining pressure during the analysis, which in other methods, in addition to the sealing function, represents the horizontal stress occurring *in situ*. It is obvious that in the case of poorly permeable rocks, such as sealing rocks, petrophysical properties are strongly dependent on the prevailing stresses. So no closing pressure on the lateral surface of the sample can result in significant distortion resulting threshold pressure (increased permeability – lower breakthrough pressure).

Apart from issues related to the mercury porosimetry, it is also problematic to determine the threshold pressure of the system of interest, and in principle the conversion of the resulting values corresponding to the target system fluids. First, based on surface tension between mercury and air and contact angle we determine the threshold pressure for the mercury. Then, in view of the fact that the surface tensions between the fluids used in the laboratory (Hg – air) and which are the subject of the analysis (in this case CO_2 – brine) are different, it is necessary to convert the results according to the relation (Busch et al 2010):

$$p_{c\,CO_2/brine}^{th} = p_{c\,Hg/air}^{th} \frac{\sigma_{CO_2/brine}\cos\theta_{CO_2/brine}}{\sigma_{Hg/air}\cos\theta_{Hg/air}}$$

Therefore, the value of contact angle (wettability) of rock with carbon dioxide is often not known, completely water-wet system is assumed ($\theta_{CO_2} = 0$). In the light of recent work (Chiquet et al., 2007), which shows that in the presence of CO_2 sealing rocks are characterized by a mixed wettability, this simplification contributes substantially to reduce the reliability of the results.

Taking into account the limitations and imperfections, the application of the presented method should be limited to a preliminary estimate sought threshold capillary pressure.

4.2 Standard approach

Direct measurement of threshold capillary pressure of seal rocks is usually carried out by displacing the wetting phase (brine) from completely saturated core by injection of nonwetting phase (gas). The most common method of measurement based on this approach is the standard method, also known as step-by-step method. The measurement can be carried out under condi-

tions corresponding to those prevailing *in situ*, and is very easy to interpret, as it is based directly on the definition of the threshold capillary pressure (Egermann et al., 2006).

During the test pressure of the nonwetting phase is raised in steps, and after each increment of pressure at the inlet of the sample (upstream pressure), behavior of the system outlet pressure (downstream pressure) is observed. Pressure is raised until the outlet pressure increases, which means that the nonwetting fluid reached the barrier of minimum capillary entry pressure, and penetrated the pore space of the analyzed rock sample. Further increase in pressure differential between the input and output of sample will result in exceeding the capillary pressure of series of the largest interconnected pore channels and nonwetting phase Darcy flow. The difference between the input pressure (nonwetting phase pressure) and the outlet pressure (wetting phase pressure) at this point is taken as the capillary breakthrough pressure for the sealing rock-nonwetting fluid system.

Accuracy of results obtained with the standard method is closely connected with the accepted values of pressure increase and the length of each step (stabilization time after each pressure increase). Pressure increment should be chosen with regard to the expected value of the threshold pressure. Too high pressure increment value will result with overstated value of the measured threshold pressure. For the test sample, for which we are not able to pre-estimate the value of P_{cth}, the study should begin "from zero". In another study of the same sample injection pressure can be started only slightly lower than the value obtained in the previous approach and use a smaller increment of pressure, so that the threshold pressure value obtained is accurate. Achieving high quality results, unfortunately, is associated with a significant investment of time, which can be considered as a fundamental drawback of the method presented.

In order to prevent fluid flow between the outer surface of the rock sample and the wall of the coreholder, the additional sealing in the form of a viton sleeve under pressure is used. Pressure applied to the lateral surface of the sample by sealing sleeve also acts as a stress that act on the sealing rock in situ.

To maximize effectiveness of carbon dioxide geological storage the storage site should provide supercritical conditions ($p > p_{c\,CO_2}$ = 73,8 bar and $T > T_{c\,CO_2}$ = 304,20 K = 31,05°C). Therefore, the laboratory measurement for the system CO_2-brine should also be conducted under such conditions. For this purpose apparatus shall be equipped with outlet pressure regulator (BPR – Back Pressure Regulator), to maintain supercritical pressure.

4.3. Continuous injection approach

This method involves the continuous injection of nonwetting phase with a constant and very low rate. With this approach, nonwetting phase pressure in the initial phase of the experiment gradually increases and the flow in the core does not occur. When injected fluid pressure exceeds the threshold value (for part of the sample closest to the inlet surface), flow in the core begins. In the case of heterogeneous samples, and these occur most frequently, the evolution of the pressure as a function of time is subject to fluctuations (increases and decreases), reflecting the diversity of the capillary threshold pressure along the axis of the core. Continuous injection method is not without drawbacks. The primary limitation is its assumption that the wetting phase viscosity gradient (water) may be omitted by using very low flow rates. As a result, such a simplification may occur to a significant revaluation of the threshold capillary pressure (Egermann et al., 2006).

5. Apparatus for standard method measurement of the capillary threshold pressure

Schematic of the apparatus is shown in Figure 2. Its main element is the high pressure Hassler-type core holder (Fig. 3). Rock sample placed in a chamber is subjected to compressive stress caused by the sealing pressure (confining pressure) achieved by injection of hydraulic fluid between the sealing viton sleeve, in which sample is placed, and steel body of core holder. Control of this pressure during the experiment is performed with a pressure gauge. In addition, the pressure chamber is equipped with connectors: inlet and outlet, supplying fluid to the rock sample through the distribution plugs. For both nozzles are mounted pressure transducers, enabling continuous pressure measurement and registration during the study. The fluid, in this case carbon dioxide, is fed to the inlet indirectly with use of pulsation-free dosing pump from the tank with a movable piston. The pump allows to adjust the pressure of the pumped fluid through the entire experiment. In order to ensure constant temperature during the study, both the core holder and the cylinder tank with piston are placed in a thermostatic chamber.

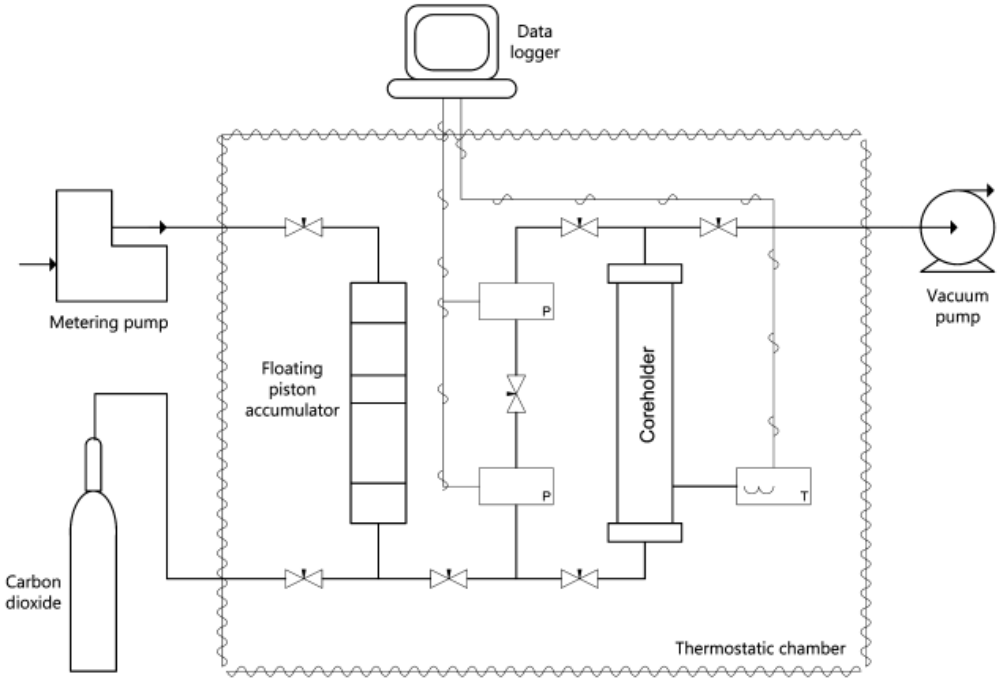


Fig. 2. Schematic of the apparatus

Fig. 3. Hassler type core holder

6. Measurements

The cores selected for the study of rock samples were cut in the form of a cylinder with a diameter of one and one and half inches in length. The extracted samples were first study of porosity using helium porosimeter. Then, permeability of samples was determined using the pressure pulse decay method. The results of these preliminary measurements are summarized in Table 1.

TABLE 1

Porosity and micro-permeability results for analyzed rock samples

Well	Depth [m]	Porosity [%]	Permeability [mD]
Plawno-1	1186,5	9,35	0,0002946
Radecin-1	916,9	19,36	0,0656041
Wierzchowo-12	1937,1	10,0	0,0015252

Prior to the main part of the experiment, rock samples were saturated with brine with total mineralization of 15 g/dm^3. This process was performed in an automatic core saturator (Vinci Technologies) under a pressure of 150 bar. Prepared samples was placed in the viton collar and then installed in the core holder. The confining pressure sealing for threshold pressure meas-

urement was set at 140 bar. After connecting the inlet of the core holder and the cylinder with piston the latter was filled with pure carbon dioxide gas stored in pressurized cylinder. During the experiment inlet pressure (CO_2 pressure) was increased in steps every ten hours. Simultaneously pressure changes on both the inlet and outlet of the core holder were recorded.

7. Results

The graphs presented below (Fig. 4-6) shows the changes of pressure during the experiment at the inlet (green) and outlet (red) from the core holder for rock samples from well Plawno-1, Radecin-1 and Wierzchowo-12.

For a sample from the well Plawno-1, due to the relatively low permeability (see Table 1), the test began with inlet pressure of 35 bar, and then every ten hours was increased by 5 bar. From the diagram (Fig. 4) can be seen that the increase in pressure at the outlet from the coreholder was recorded when the inlet pressure was increased from the 55.8 to 61.2 bar.

Measurement for sample from the well Radecin-1, with the highest porosity and permeability from among the tested samples, was started at 6.8 bar pressure. The pressure increase downstream (Fig. 5) was observed after a relatively short time after increasing pressure from 22.7 to 29.8 bar.

For a sample from well Wierzchowo-12 (Fig. 6) changes in pressure at the core holder outlet were recorded when the inlet pressure increased from 43.1 to 50.3 bar.

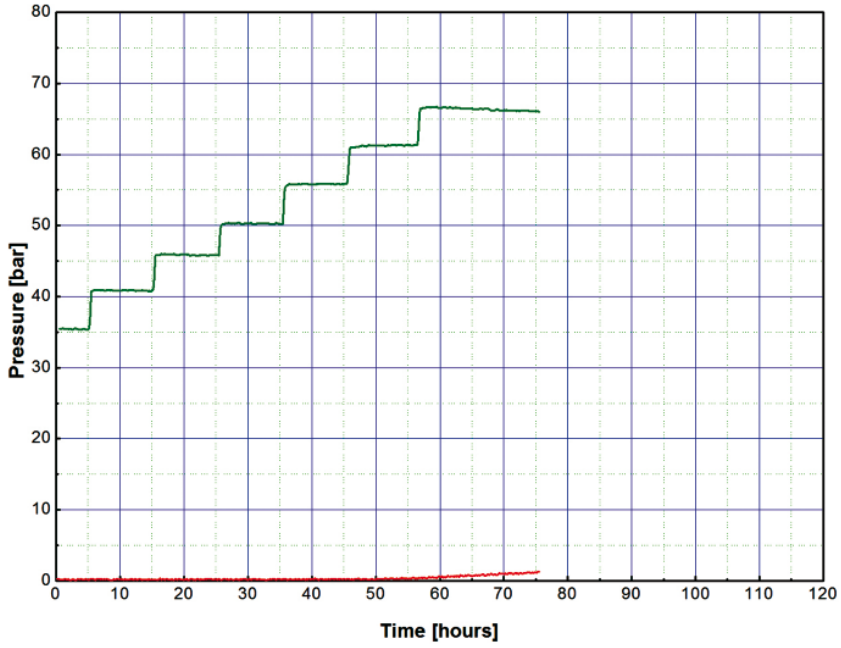


Fig. 4. Inlet (green) and outlet (red) pressure changes for Plawno-1 rock sample

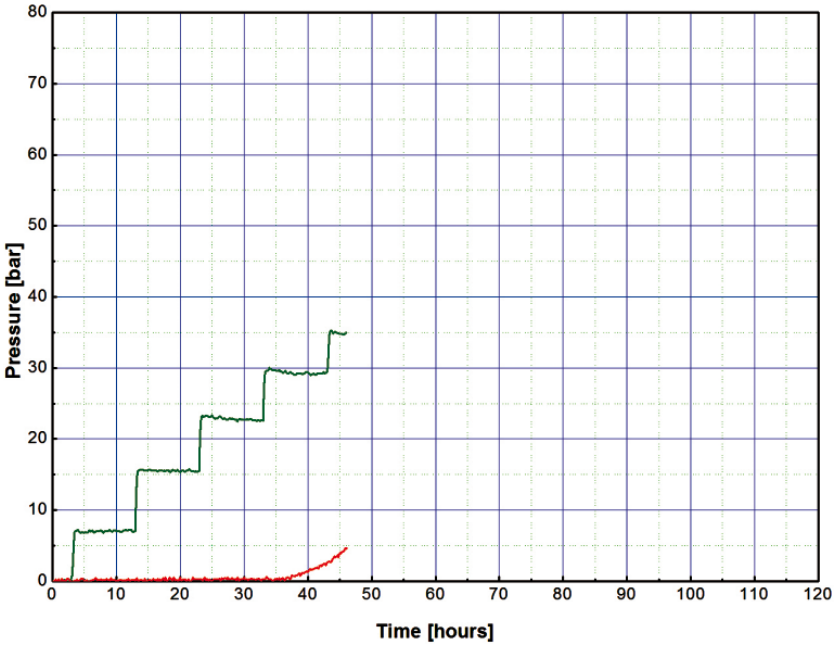
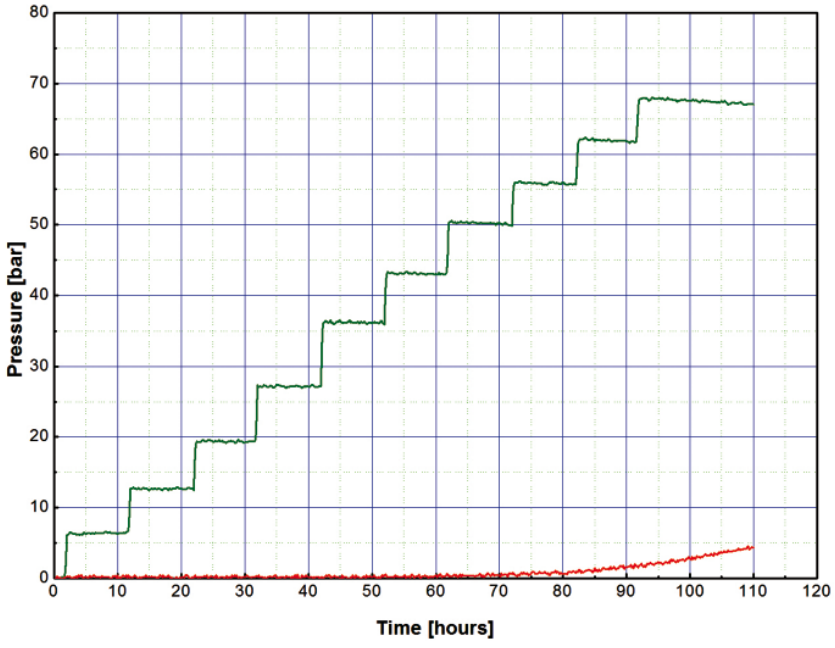


Fig. 5. Inlet (green) and outlet (red) pressure changes for Radecin-1 rock sample



Rys. 6. Inlet (green) and outlet (red) pressure changes for Wierzchowo-12 rock sample

8. Conclusions

Values of the capillary threshold pressure for a given rock sample were estimated and placed in a certain range of values, and so for the sample Plawno-1 the range is of 55.8÷61.2 bar, for Radecin-1 interval 22.7÷29.8 bar and for the Wierzchowo-12 sample range 43.1÷50.3 bar. In order to obtain accurate values of the threshold pressure it is necessary to perform a re-examination of each sample within the obtained interval of threshold pressure, with an appropriately small increments of growth over a prolonged period of time steps.

First results obtained with new apparatus seem to be promising and are a good base for further work, as for example incorporating the back pressure regulator for providing supercritical conditions. Planned are also measurements of water (brine) relative permeability after mounting the core in the apparatus and before injecting CO_2, and relative permeability for CO_2 after breakthrough.

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